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OBSERVED FRACTIONATION IN GROUND LEVEL FALLOUT FROM THREE NUCLEAR CRATERING DETONATIONS

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Abstract

Data relating to radionuclide deposition (fallout) within a few miles of the Danny Boy, Sedan, and Palanquin nuclear cratering shots are examined for evidence of fractionation - i.e., differences in relative proportions of fission products in debris samples as compared with the relative proportions originally formed. A fractionation index is computed for several fission-product mass chains produced in each event. This index is particularly useful because it is independent of the total yield and fission yield of the detonation and also of the total amount of radioactive material deposited in the early fallout pattern. By comparing these indices with one another, one can determine

whether fractionation occurred and obtain a quantitative estimate of its degree. For the three events studied here, only Danny Boy showed unambiguous evidence of fractionation in the early fallout, and the degree of fractionation was small. In Danny Boy there was only a factor of four difference between most enriched and most depleted species, as compared with the factors of several hundred that have been observed in many late time samples of airborne debris. If this small amount of fractionation proves to be true in general for cratering shots, then predictions of early-fallout gamma radiation patterns will be greatly simplified.

Introduction

The radioactive products of a nuclear explosion are said to have undergone fractionation if their relative proportions in samples taken at various locations differ significantly from their relative proportions as formed. This report describes a study of fractionation in the early fallout from the nuclear cratering shots Danny Boy, Sedan, and Palanquin. Published fallout data for these shots was the

basic information used in the study. A normalization procedure was applied to the published data as follows: the amount of each radionuclide (or mass chain) of interest measured on a fallout tray is related to the gamma radiation exposure rate measured at the tray location and to the amount of that radionuclide produced per kiloton of fission by the device. The result is an index number for each

radionuclide of interest. These "fractionation indices" show the degree of fractionation that has occurred in the deposited early fallout debris.

This technique has the advantage of reducing a large number of individual measurements to a meaningful, manageable, and comprehensible set of fractionation index numbers. It is useful and illuminating because the quantitative measure of ground deposition for a particular mass chain (or a specific nuclide in

the case of a shielded nuclide) can be compared directly to the ambient gamma radiation field. The index allows a comparison of the deposition of the various mass chains one to another, and it enables the deposition from one cratering event to be contrasted with that from another on a mass-chain-by-mass-chain basis. Furthermore, the method is independent of fission yield, total yield, and total amount of radioactive material deposited in the close-in fallout patterns.

The Fractionation Index

The fractionation index I_i is calculated for each measured fission product at each tray location in the fallout field as follows:

$$I_i = \frac{\text{No. atoms in tray}}{\text{area of tray}}$$

$$\times \frac{1}{\text{No. atoms from 1 kt fission}}$$
 $\times \frac{1}{\text{gamma exposure rate at tray}}$

or, symbolically,

$$I_i = \frac{M_i}{A_{tr}} \times \frac{1}{N_i} \times \frac{1}{(d\gamma/dt)_{H+1 \text{ hr}}}$$
,

where

I = index for mass chain i as represented by nuclide i,

M_i = atoms of nuclide i measured in tray, corrected to zero time,

 A_{tr} = area of tray in ft^2 ,

N_i = atoms of nuclide i produced by 1 kt of fission, corrected to zero time (in practice, usually the total yield of chain i per kiloton, expressed as number of atoms),

 $(d\gamma/dt)_{H+1 \text{ hr}}$ = gamma exposure rate in R/hr, measured with a 4π detector 3 ft above surface at tray location and decaycorrected to a time of H + 1 hr.

Since the gamma exposure rate at a particular location is directly proportional to the area concentration of gamma-emitting nuclides in the vicinity, one notes that, in the absence of fractionation, the index will have the same value for all nuclides at all locations. When fractionation takes place, the index gives a quantitative measure of the enrichment or depletion of a mass chain relative to the total assemblage. If present, neutron-induced

gamma-emitting nuclides will add to the exposure rate and thus decrease the magnitude of the index. This consideration is of interest when comparing different

cratering events. The utility of this index is demonstrated below by examining its application to the cratering events Danny Boy, Sedan, and Palanquin.

Sources of the Data

The report of Miskel and Bonner was the source of Danny Boy information. They used measurements from nine tray locations extending from 2,500 to 25,000 ft from ground zero and studied 11 fission product nuclides. The gamma radiation exposure rates measured at the tray locations varied by more than a factor of 100 with distance from ground zero, as did the radiochemical determinations of each nuclide expressed as disintegrations per minute per square foot. Miskel and Bonner divided the latter by the former to give a factor designated as J_i. While the $J_{\underline{i}}$ for any one nuclide was not exactly constant with distance from ground zero, the values were reasonably consistent and, furthermore, no discernible trend with distance was apparent. Also, the ratios of J_i values between different nuclides showed no trend with distance. Thus it was concluded that there was no change in fractionation over the distances involved, and so the determinations for the various trays at different distances were averaged.

Information concerning Sedan was obtained from the report by Lane. 2 He reports equivalent fissions per gram for material from fallout trays between 5,800 and 19,200 ft from ground zero for a series of nuclides. The determinations show random scatter and do not indicate a trend with distance. Therefore, the values for different trays were averaged. A report by Nordyke and Williamson³ provided the experimental determination of fallout-mass area density divided by gamma field readings in units of (kg/m²) /(R/hr) over the same fallout area. These two sources provided the necessary input to calculate the fractionation indices for

Schwartz⁴ provided the early fallout field tray information for Palanquin. He reported a fraction of total atoms produced per ft² divided by the field exposure rate at the tray locations in R/hr at H + 1 hr for the different nuclides studied. The trays were placed from about 2,500 to 25,000 ft from ground zero; the data show no trend with distance so the determinations for the different trays were averaged.

Calculated Fractionation Indices

Table I gives the fractionation indices obtained for the various mass chains in

these three cratering events. The mass chains are divided into the conventional

Table I. Calculated fractionation indices for early fallout debris from the Danny Boy, Sedan, and Palanquin nuclear cratering shots.

| Group | Nuclide | Index (units of 10^{-11} kt-hr/ft ² -R) | | |
|--------------|----------------------|--|-------|-----------|
| | | Danny Boy | Sedan | Palanquin |
| Volatile | Sr ⁸⁹ | 1.46 | 0.81 | 0.30 |
| | Sr ⁹⁰ | 1.94 | 1.26 | |
| | ${ m Y}^{91}$ | | 1.11 | - |
| | ${ m Te}^{132}$ | ************************************** | 0.77 | 1.04 |
| | Cs^{137} | 1.72 | 0.59 | 0.35 |
| | Ba ¹⁴⁰ | 1.21 | 0.98 | 0.74 |
| Intermediate | Ru ¹⁰³ | | 0.54 | |
| | $\frac{100}{Ru}$ 106 | | 0.71 | |
| | I^{131} | | 0.95 | 0.58 |
| | Cs^{136} | 1.57 | 0.99 | 1.12 |
| | Ce^{141} | 0.90 | 1.12 | 0.88 |
| Nonvolatile | ${ m Zr}^{95}$ | 0.38 | 1.19 | 0.72 |
| | $^{ m Mo}^{99}$ | 0.42 | 0.91 | 0.79 |
| | Ce^{144} | 0.38 | 1.13 | 0.62 |
| | Nd^{147} | 0.37 | _ | 0.69 |
| | Eu ¹⁵⁶ | 0.34 | | 0.78 |

three groups: volatile, intermediate, and nonvolatile.

For Danny Boy the indices are roughly four times as high for the volatiles as for the nonvolatiles, while the intermediates fall between these extremes. This clearcut, systematic difference in the indices is taken as evidence that fractionation occurred in the Danny Boy early fallout, but to a much smaller degree (factor of four) than might have been predicted on the basis of other fractionation studies. (For example, late time samples of airborne debris have shown fractionation involving factors of several hundred.)

For Sedan the indices show little or no evidence of fractionation. They scatter

somewhat, but not greatly for this type of information.

For Palanquin the indices suggest that some slight fractionation is present. Since the nonvolatile group forms such a consistent set, one is tempted to infer that the ${\rm Sr}^{89}$ and ${\rm Cs}^{137}$ mass chains are depleted and that all other nuclides are present in just about their production proportions.

As a rough check on the magnitude of the indices in Table I, a theoretical estimate of the fractionation index for unfractionated fission products can be calculated as follows. The index has the units $kt-hr/ft^2-R$; if we use the most probable value of 2500 R/hr for 1 kt/mi²

over Nevada Test Site terrain for unfractionated fission products, we obtain (since $1 \text{ mi}^2 = 2.78 \times 10^7 \text{ ft}^2$)

$$I = \frac{1}{2.78 \times 10^7 \times 2.5 \times 10^3} = 1.44 \times 10^{-11}.$$

This independently calculated theoretical value for the unfractionated index agrees closely with the experimental indices in Table I, thus giving support to their validity.

The relatively small spread of the numbers in Table I (about a factor of six from smallest to largest) is remarkable in view of the considerable differences in venting phenomenology among the three

shots and the variety of sources for the radiochemical data used. Danny Boy data was supplied by the U.S. Air Force; Sedan data by Tracerlab, Nuclear Science and Engineering Corp., and Hazleton Nuclear Science Corp.; and Palanquin data by Lawrence Radiation Laboratory, Livermore.

To summarize, there appears to have been little fractionation in the early fall-out from these three cratering events, with Danny Boy being the only one of the three to show unambiguous evidence of fractionation. If this observation proves to be true in general for cratering shots, then predictions of early-fallout gamma radiation patterns will be greatly simplified.

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